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PATENT ABSTRACTS OF JAPAN

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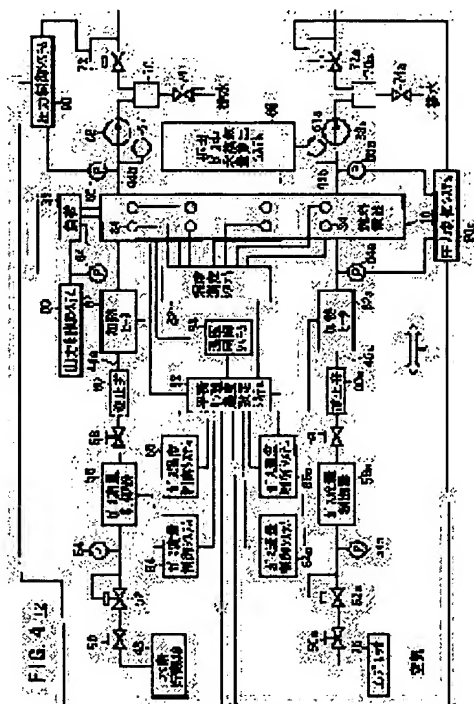
ISE MASAHIRO

(54) OPERATING METHOD OF PHOSPHORIC ACID FUEL CELL

(57)Abstract:

PROBLEM TO BE SOLVED: To enable to operate in good condition the phosphoric acid fuel cell in low temperature without using additional equipment.

SOLUTION: In operating the phosphoric acid fuel cell 10 under the condition in which the reaction formed water exists in liquid state, the operating condition is established so that the phosphoric acid concentration, in which the quantity of the reaction formed water lowering the phosphoric acid concentration and the quantity of the water vaporizing from the phosphoric acid are equalized, becomes same or higher than the standard phosphoric acid concentration in which it can maintain the desired performance. Specifically, a gas flow control system 84, 84a, a gas temperature control system 86, 86a and a pressure control system 90, 90a are controlled as required by the equalizing phosphoric acid concentration setting system 92, and the performance deterioration and generation performance degradation of the phosphoric acid fuel cell are effectively avoided.



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CLAIMS

[Claim(s)]

[Claim 1] It has a zygote which consists of an anode lateral electrode and a cathode lateral electrode on both sides of an electrolyte to which impregnation of the phosphoric acid was carried out. While supplying fuel gas to said anode lateral electrode through fuel gas passage It is the operating method of a phosphoric acid fuel cell which supplies oxidizer gas to said cathode lateral electrode through an oxidizer gas passageway. Phosphoric-acid concentration at the time of an amount of said produced water to which phosphoric-acid concentration is reduced, and an amount of water which evaporates out of a phosphoric acid balancing, in case it operates under conditions in which produced water exists as liquefied water An operating method of a phosphoric acid fuel cell characterized by setting up operating conditions so that it may become more than criteria phosphoric-acid concentration which can maintain desired engine performance.

[Claim 2] An operating method of a phosphoric acid fuel cell characterized by controlling said phosphoric-acid concentration in an operating method according to claim 1 more than said criteria phosphoric-acid concentration by setting up a fuel gas utilization factor and a rate of oxidizer gas utilization.

[Claim 3] An operating method of a phosphoric acid fuel cell characterized by controlling said phosphoric-acid concentration in an operating method according to claim 1 or 2 more than said criteria phosphoric-acid concentration by setting up operating pressure by the side of said fuel gas, and operating pressure by the side of said oxidizer gas.

[Claim 4] An operating method of a phosphoric acid fuel cell characterized by controlling said phosphoric-acid concentration in an operating method given in claim 1 thru/or any 1 term of 3 more than said criteria phosphoric-acid concentration by setting temperature by the side of said fuel gas, and temperature by the side of said oxidizer gas as an elevated temperature rather than an operating temperature.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] While this invention has the zygote which consists of an anode lateral electrode and a cathode lateral electrode on both sides of the electrolyte to which impregnation of the phosphoric acid was carried out and supplies fuel gas to said anode lateral electrode through a fuel gas supply system, it relates to the operating method of the phosphoric acid fuel cell which supplies oxidizer gas to said cathode lateral electrode through an oxidizer gas supply system.

[0002]

[Description of the Prior Art] The phosphoric acid fuel cell (PAFC) which is one gestalt of a fuel cell The phosphoric acid which is a liquid electrolyte on for example, both sides of the electrolyte matrix layer which carried out impregnation to poly membranes, such as polybenzimidazole The anode lateral electrode and cathode lateral electrode which make carbon a subject, respectively an opposite the electrolyte and electrode zygote *(ed) and constituted It has the generation-of-electrical-energy cel (unit fuel cell cel) constituted by pinching with a separator (bipolar board), and only a predetermined number carries out the laminating of this generation-of-electrical-energy cel, and is usually using it as a fuel cell stack.

[0003] In this kind of fuel cell, hydrogen is ionized on a catalyst electrode and the fuel gas supplied to the anode lateral electrode, for example, the gas which mainly contains hydrogen, (hydrogen content gas) moves to a cathode lateral electrode side through an electrolyte. The electron produced in the meantime is taken out by the external circuit, and is used as electrical energy of a direct current. In addition, since oxidizer gas, for example, the gas which mainly contains oxygen, or air (oxygen content gas) is supplied, a hydrogen ion, an electron, and oxygen react in this cathode lateral electrode, and water is generated by the cathode lateral electrode.

[0004] In the above-mentioned phosphoric acid fuel cell, the operating temperature is comparatively set as the elevated temperature (before or after 120 degrees C - 190 degrees C), and generally, said phosphoric acid fuel cell is warmed using heating means, such as a heater, until it results in steady operation. And after temperature up of this phosphoric acid fuel cell is carried out to 100 degrees C or more of abbreviation, it carries out temperature up to 120 degrees C - 190 degrees C by operating said phosphoric acid fuel cell using the self-pyrexia accompanying a generation of electrical energy.

[0005]

[Problem(s) to be Solved by the Invention] By the way, while the ratio in which the produced water accompanying a generation of electrical energy of a phosphoric acid fuel cell exists as liquefied water becomes high like [at the time of a start up] in the bottom of a comparatively low temperature of less than 100 degrees C, it has been [steam] hard coming-izing [this liquefied water]. For this reason, the liquefied water with which liquefied water reduces the concentration of a phosphoric acid, makes the amount of phosphoric acids increase, and exceeds the amount which can be accumulated in an anode lateral electrode and a cathode lateral electrode will exist.

[0006] Thus, if a phosphoric acid overflows into an anode lateral electrode and a cathode lateral electrode, the phosphoric acid currently held in the electrolyte matrix layer will flow out, and this phosphoric acid that flowed out will be discharged by the exterior of the main part of a phosphoric acid fuel cell through fuel gas passage and an oxidizer gas passageway. When it is made to reboot by this after suspending operation of a phosphoric acid fuel cell, the cell engine performance cannot maintain the initial engine performance, but the problem that degradation is caused is pointed out.

[0007] furthermore, if a phosphoric acid overflows into an anode lateral electrode and a cathode lateral electrode, the activity of the catalyst which has the function to promote a reaction required for a generation of electrical energy falls, or fuel gas passage and an oxidizer gas passageway blockade -- having -- reactant gas (fuel gas and/or oxidizer gas) --

circulating -- being hard -- there is a problem that the generation-of-electrical-energy engine performance falls.

[0008] Then, although it is possible to use the phosphoric-acid makeup equipment according to individual in order to improve the performance degradation accompanying runoff of a phosphoric acid, there is a problem that the whole fuel cell system will be enlarged.

[0009] Moreover, in order to carry out temperature up of the phosphoric acid fuel cell promptly to 100 degrees C or more, it is possible to use a large-sized heater, but similarly, while the whole fuel cell system is enlarged, the problem of not being economical is pointed out.

[0010] This invention does not solve this kind of problem, and equipment is not enlarged, and it is an easy configuration and control and aims at offering the operating method of the phosphoric acid fuel cell which can prevent the degradation by produced water certainly.

[0011]

[Means for Solving the Problem] In an operating method of a phosphoric acid fuel cell concerning this invention, in case it operates under conditions in which produced water exists as liquefied water, operating conditions are set up so that phosphoric-acid concentration at the time of an amount of said produced water to which phosphoric-acid concentration is reduced, and an amount of water which evaporates out of a phosphoric acid balancing may become more than criteria phosphoric-acid concentration which can maintain desired engine performance.

[0012] Usually, a phosphoric acid used for a phosphoric acid fuel cell carries out melting of 5 oxidation 2 Lynn to water, especially a high-concentration phosphoric acid used for a phosphoric acid fuel cell has high hygroscopicity, and it tends [very] to mix it with water. In this case, although steamy components of a phosphoric acid are 5 oxidation 2 Lynn (accuracy dimer) and water, vapor pressure of 5 oxidation 2 Lynn is very low to near 200 degree C. For this reason, although a steamy component of a phosphoric acid is ****, vapor pressure of a phosphoric acid differs from vapor pressure of water.

[0013] Temperature of a phosphoric acid and relation of maximum vapor tension to phosphoric-acid concentration are shown in a diagram of drawing 1, and, specifically, a diagram of 0% of phosphoric-acid concentration in drawing 1 is equivalent to a maximum-vapor-tension diagram of water. Thereby, while water in a phosphoric acid becomes easy to evaporate so that phosphoric-acid concentration is low, it has stopped easily maximum vapor tension of a phosphoric acid changing with phosphoric-acid concentration a lot, and being able to evaporate, so that phosphoric-acid concentration is high.

[0014] In a phosphoric acid fuel cell, in the time of low temperature like [at the time of starting], or low-power output operational status, when temperature of said phosphoric acid fuel cell usually falls rather than operating temperature (before or after 120 degrees C - 190 degrees C), a vapor rate of water in a phosphoric acid becomes slow, and will be in a condition with many amounts of generation water from an amount of evaporating water. Therefore, while phosphoric-acid concentration in an electrolyte and an electrode zygote becomes low, maximum vapor tension of a phosphoric acid becomes high, and water becomes easy to evaporate.

[0015] For this reason, an amount of generation water in which phosphoric-acid concentration falls to according to an amount of generation water, and phosphoric-acid concentration is reduced at a certain event, and an amount of water which evaporates out of a phosphoric acid reach a balance. In this condition, the amount of phosphoric acids is increasing, and when it increases more than the amount of phosphoric acids in which this amount of phosphoric acids is stored in an electrolyte and an electrode zygote, there is a possibility that a phosphoric acid may be discharged by the exterior of a phosphoric acid fuel cell from fuel gas passage and an oxidizer gas passageway, and performance degradation may arise.

[0016] If operating conditions are set up so that phosphoric-acid concentration at the time of an amount of produced water to which phosphoric-acid concentration is reduced, and an amount of water which evaporates out of a phosphoric acid balancing by this may become more than criteria phosphoric-acid concentration which deterioration of engine performance including generation-of-electrical-energy engine performance does not produce, even if it is the case that temperature of a phosphoric acid fuel cell is low, performance degradation will not be caused and an efficient generation of electrical energy will become executable.

[0017] Moreover, phosphoric-acid concentration which performance degradation does not produce is influenced by an internal configuration, a size, etc. in an electrolyte and an electrode zygote, and is the thing of a proper at the phosphoric acid fuel cell. Therefore, it is necessary to set up beforehand experimentally phosphoric-acid concentration which can maintain desired engine performance.

[0018] The amount m (mol/min) of water in which an active electrode area reduces an amount of water generated by per unit time amount when it is S (cm²), i.e., concentration of a phosphoric acid, by I (A/cm²) has fixed programmed-current density of operating conditions regardless of the fuel gas utilization factor ra (%) and the rate rc of oxidizer gas

utilization (%), and it asks by the following (1) type.

[0019]

[Equation 1]

$$m = I \times S \times \frac{60}{96500 \times 2} \dots\dots(1)$$

[0020] Ta^{**} and oxidizer gas temperature Next, Tc^{**} , [an operating temperature] [T degrees C and fuel gas temperature] Pa (kPa) and oxidizer side operating pressure Pc (kPa), [fuel gas side operating pressure] The supply flow rate (fuel gas flow rate) of fuel gas fa (normal l/min) (However, normal l/min is 0 degree C which is reference condition, and the flow rate converted into 1atm) The supply flow rate (oxidizer quantity of gas flow) of oxidizer gas fc (normal l/min), The quantity of gas flow discharged from a fuel gas side fea (normal l/min), The quantity of gas flow discharged from an oxidizer gas side sets to 1-a (%) the ratio which evaporates the ratio which evaporates from the inside of the amount of the water in which the concentration of fec (normal l/min) and a phosphoric acid is reduced, and a fuel gas side from an a (%) and oxidizer gas side.

[0021] The quantity of gas flow fea discharged from a fuel gas side is the sum with the water vapor content which evaporates from the inside of the flow rate of the fuel gas which was not consumed by generation of electrical energy, and the amount of the water in which the concentration of a phosphoric acid is reduced, and a fuel gas side (it asks from the following (2) types).

[0022]

[Equation 2]

$$fea = fa \times \frac{100 - ra}{100} + m \times \frac{a}{100} \times 22.4 \dots\dots(2)$$

[0023] Then, the maximum vapor tension Ps and a of the phosphoric acid in the blowdown fuel gas in the event of the amount of the water with which fuel gas temperature Ta^{**} and oxidizer gas-temperature Tc^{**} evaporate out of the amount of the generation water in which phosphoric-acid concentration is reduced, and a phosphoric acid when equal to the operating temperature of T degrees C being balancing is called for by the following (3) types.

[0024]

[Equation 3]

$$\begin{aligned} Ps, a &= \frac{m \times \frac{a}{100} \times 22.4}{fea} \times Pa \\ &= \frac{m \times \frac{a}{100} \times 22.4}{fa \times \frac{100 - ra}{100} + m \times \frac{a}{100} \times 22.4} \times Pa \dots\dots(3) \end{aligned}$$

[0025] On the other hand, the quantity of gas flow fec discharged from an oxidizer gas (it is air and nitrogen is 79wt (s)% at 21wt(s)% for oxygen) side is calculated from the following (4) types.

[0026]

[Equation 4]

$$fec = fc \times (0.21 \times \frac{100 - rc}{100} + 0.79) + m \times \frac{100 - a}{100} \times 22.4 \dots\dots(4)$$

[0027] Furthermore, the maximum vapor tension Ps and c of the phosphoric acid in blowdown oxidizer gas is similarly called for from the following (5) types.

[0028]

[Equation 5]

$$\begin{aligned} Ps, c &= \frac{m \times \frac{100 - a}{100} \times 22.4}{fec} \times Pc \\ &= \frac{m \times \frac{100 - a}{100} \times 22.4}{fc \times (0.21 \times \frac{100 - rc}{100} + 0.79) + m \times \frac{100 - a}{100} \times 22.4} \times Pc \dots\dots(5) \end{aligned}$$

[0029] Moreover, the maximum vapor tension of a phosphoric acid and the relation of phosphoric-acid concentration are shown in the diagram of drawing 2, and the phosphoric-acid concentration in the temperature of T degrees C is called for as a function of the maximum vapor tension of a phosphoric acid. In the fixed operating temperature of T degrees C, phosphoric-acid concentration is high, so that the maximum vapor tension of a phosphoric acid is low, so that it may be understood from drawing 2. Therefore, it can ask for the phosphoric-acid concentration to which the amount of the generation water in which phosphoric-acid concentration is reduced in the operating temperature of T degrees C, and the amount of the water which evaporates out of a phosphoric acid are balancing using the maximum vapor tension P_s and a of the phosphoric acid in blowdown fuel gas, and the maximum vapor tension P_s and c of the phosphoric acid in oxidizer gas, respectively.

[0030] Specifically, the maximum vapor tension P_s and a of the phosphoric acid in blowdown fuel gas is the function of the quantity of gas flow f_{ea} discharged from an operating pressure [by the side of fuel gas] P_a , and fuel gas side. The above-mentioned blowdown fuel gas flow rate f_{ea} is the function of the amount m of the water in which the concentration of a phosphoric acid is reduced, and the fuel gas utilization factor r_a , and said its blowdown fuel gas flow rate f_{ea} increases, so that the fuel gas utilization factor r_a is [m] low at the fixed time. Therefore, the maximum vapor tension P_s and a of the phosphoric acid in blowdown fuel gas becomes low, when the operating pressure P_a by the side of fuel gas is low, or when the fuel gas utilization factor r_a is low, consequently phosphoric-acid concentration becomes high.

[0031] Similarly, the maximum vapor tension P_s and c of the phosphoric acid in blowdown oxidizer gas is the function of 0 degree C which is the operating pressure P_c , the rate r_c of oxidizer gas utilization, and reference condition by the side of oxidizer gas, and the oxidizer quantity of gas flow f_c converted into 1 atm. Therefore, the maximum vapor tension P_s and c of the phosphoric acid in blowdown oxidizer gas becomes low, when the operating pressure P_c by the side of oxidizer gas is low, or when the rate r_c of oxidizer gas utilization is low, consequently phosphoric-acid concentration becomes high.

[0032] By this by setting up the fuel gas utilization factor r_a , the rate r_c of oxidizer gas utilization and/or the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas The phosphoric-acid concentration to which the amount of the generation water in which phosphoric-acid concentration is reduced, and the amount of the water which evaporates out of a phosphoric acid are balancing turns into more than the criteria phosphoric-acid concentration that does not cause performance degradation, namely, can maintain the desired engine performance, and can prevent degradation certainly also in the time of low-temperature operation.

[0033] On the other hand, phosphoric-acid concentration is called for as a function of the maximum vapor tension of a phosphoric acid, and temperature (refer to drawing 2). For this reason, if the temperature to which the amount of the generation water in which phosphoric-acid concentration is reduced, and the amount of the water which evaporates out of a phosphoric acid are balancing is highly set up when the maximum vapor tension of a phosphoric acid is fixed, it will become possible to make high phosphoric-acid concentration at the time of a balance.

[0034] That is, by making fuel gas temperature T_a^{**} and oxidizer gas-temperature T_c^{**} higher than the operating temperature of T degrees C, phosphoric-acid temperature T^{**} in the balanced interface at the time of the amount of the generation water in which phosphoric-acid concentration is reduced, and the amount of the water which evaporates out of a phosphoric acid being balancing can be made higher than the operating temperature of T degrees C, and phosphoric-acid concentration at the time of a balance can be made high. In addition, as for the relation between fuel gas temperature T_a^{**} and oxidizer gas-temperature T_c^{**} , and phosphoric-acid temperature T^{**} in a balanced interface, it is desirable to be the thing of a proper and to ask the phosphoric acid fuel cell experimentally.

[0035]

[Embodiment of the Invention] Drawing 3 is configuration explanatory drawing of the phosphoric acid fuel cell 10 for enforcing the operating method concerning the operation gestalt of this invention, and drawing 4 is the outline block diagram of the fuel cell system 12 by which said phosphoric acid fuel cell 10 is incorporated.

[0036] A phosphoric acid fuel cell 10 is equipped with an electrolyte and the electrode zygote 14, the separators 16a and 16b that are the bipolar boards arranged to both sides of this electrolyte and electrode zygote 14, and the electrodes 18a and 18b for current collection arranged on the outside of said separators 16a and 16b as shown in drawing 3. end plates 20a and 20b are arranged, and a phosphoric acid fuel cell 10 is constituted by bolting ***** in one with the bolt which said end plates 20a and 20b do not illustrate on the outside of the electrodes 18a and 18b for current collection.

[0037] An electrolyte and the electrode zygote 14 are equipped with the electrolyte matrix layer 22 which carried out impregnation of the phosphoric acid to poly membranes, such as basic polymer, for example, a polybenzimidazole film etc., and the anode lateral electrode 26 and the cathode lateral electrode 28 opposite-**(ed) by both sides of said electrolyte matrix layer 22 through the rim-of-duct-mouths-like spacer 24.

[0038] The fuel gas passage 30 and the oxidizer gas passageway 32 are formed in the field which counters the anode lateral electrode 26 and the cathode lateral electrode 28 at Separators 16a and 16b. Corresponding to the interface of the anode lateral electrode 26 and the cathode lateral electrode 28, two or more temperature sensors 34 are arranged at Separators 16a and 16b.

[0039] As shown in drawing 4, the loads 36, such as a motor, are connected to the electrodes 18a and 18b for current collection which constitute a phosphoric acid fuel cell 10. Fuel gas supply path 44a and fuel gas blowdown path 44b which are open for free passage to the fuel gas passage 30, and oxidizer gas supply path 46a and oxidizer gas blowdown path 46b which are open for free passage to the oxidizer gas passageway 32 are connected to a phosphoric acid fuel cell 10.

[0040] The source 48 of hydrogen storage for supplying from the upstream the hydrogen content gas which is fuel gas with high voltage toward the entrance side of a phosphoric acid fuel cell 10, a solenoid valve 50, a reducing valve 52, a pressure sensor 54, the quantity-of-gas-flow controller 56, the isolation valve 58, the check valve 60, the heating heater 62, and the input pressure sensor 64 are arranged at fuel gas supply path 44a. While the output-pressure sensor 66, the dew-point sensor 67, a heat exchanger 68, the vapor-liquid-separation machine 70, and a back-pressure valve 72 are arranged at fuel gas blowdown path 44b, the solenoid valve 74 is formed in the outlet side of this vapor-liquid-separation machine 70.

[0041] Oxidizer gas supply path 46a and oxidizer gas blowdown path 46b are constituted like the above-mentioned fuel gas supply path 44a and fuel gas blowdown path 44b. Sign a is given to the same component at the same reference numeric character, and the detailed explanation is omitted. The compressor 76 for supplying air as oxidizer gas is arranged at the upstream of this oxidizer gas supply path 46a.

[0042] While the output-control system 80 is connected to a load 36, two or more temperature sensors 34 built into the phosphoric acid fuel cell 10 are connected to the thermometry system 82. While the quantity-of-gas-flow control systems 84 and 84a are connected to the quantity-of-gas-flow controllers 56 and 56a, the gas-temperature control systems 86 and 86a are connected to the heating heaters 62 and 62a. The amount calculation system 88 of emission gas Nakamizu steams is connected to the dew-point sensors 67 and 67a, and pressure control systems 90 and 90a are connected to the input pressure sensors 64 and 64a, the output-pressure sensors 66 and 66a, and back-pressure valves 72 and 72a.

[0043] While the thermometry system 82 is connected to the balanced phosphoric-acid concentration setting-out system 92, a load 36, the quantity-of-gas-flow control systems 84 and 84a, the gas-temperature control systems 86 and 86a, the amount calculation system 88 of emission gas Nakamizu steams, pressure control systems 90 and 90a, and a climate control system 94 are connected to this balanced phosphoric-acid concentration setting-out system 92.

[0044] Thus, actuation of the fuel cell system 12 incorporating the phosphoric acid fuel cell 10 constituted is explained below.

[0045] After high-pressure hydrogen content gas is supplied from the source 48 of hydrogen storage and the pressure of the maximum of operating pressure decompresses through a reducing valve 52, the rate of gas utilization is controlled by the fuel gas supply path 44a side by the quantity-of-gas-flow controller 56. After temperature up of this hydrogen content gas is carried out to predetermined temperature at the heating heater 62, by controlling a back-pressure valve 72, it is adjusted to predetermined operating pressure and introduced into the fuel gas passage 30 established in separator 16a by the side of the anode lateral electrode 26 which constitutes a phosphoric acid fuel cell 10.

[0046] The hydrogen content gas supplied to the fuel gas passage 30 diffuses the hydrogen gas of the amount specified at the rate of gas utilization in the anode lateral electrode 26, is consumed by catalytic reaction within a catalyst bed, and contributes to a generation of electrical energy. The remaining portion of hydrogen content gas is discharged at the fuel gas blowdown path 44b side including the amount discharged from the inside of the generation water in which phosphoric-acid concentration is reduced, and a fuel gas side.

[0047] On the other hand, in the oxidizer gas supply path 46a side, after oxygen content gas, for example, air, is high-voltage-ized through a compressor 76, the pressure of the maximum of operating pressure decompresses by reducing-valve 52a. After the rate of gas utilization is controlled through quantity-of-gas-flow controller 56a, temperature up of this air is carried out to laying temperature by heating heater 62a.

[0048] Furthermore, by controlling back-pressure valve 72a, air is adjusted to predetermined operating pressure and introduced into the oxidizer gas passageway 32 currently formed in separator 16b by the side of the cathode lateral electrode 28 of a phosphoric acid fuel cell 10. The amount specified at the rate of gas utilization is spread in the cathode lateral electrode 28, is consumed by catalytic reaction within a catalyst bed, and contributes the air supplied to the oxidizer gas passageway 32 to a generation of electrical energy. The remaining portion of this air is discharged by oxidizer gas blowdown path 46b through the oxidizer gas passageway 32 including the amount discharged from the

inside of the generation water in which phosphoric-acid concentration is reduced, and an oxidizer gas side.

[0049] In this case, a required current value is set up from the demand output in that event by the output-control system 80, and the amount of electrons passed to the closed circuit which consists of a phosphoric acid fuel cell 10 and a load 36 based on this current value is determined. And the amount of the fuel gas consumed according to the amount of electrons passed to a closed circuit and oxidizer gas is determined, and the amount m of the generation water in which phosphoric-acid concentration is reduced (mol/min) is determined.

[0050] Next, the dew-point of the fuel gas discharged and oxidizer gas is detected through the dew-point sensors 67 and 67a connected to the outlet side of a phosphoric acid fuel cell 10. This dew-point data is sent to the amount calculation system 88 of emission gas Nakamizu steams, and the moisture from the phosphoric acid discharged at a moisture [from the phosphoric acid discharged at a fuel gas side] and oxidizer gas side is computed in this amount calculation system 88 of emission gas Nakamizu steams. Furthermore, ratio a (%) which evaporates from the inside of the amount of the water in which the concentration of a phosphoric acid is reduced, and a fuel gas side is computed, and this ratio a (%) is sent to the balanced phosphoric-acid concentration setting-out system 92.

[0051] On the other hand, the operating temperature of a phosphoric acid fuel cell 10 is detected by the thermometry system 82 through two or more temperature sensors 34 arranged in this phosphoric acid fuel cell 10, and the minimum value of this detected temperature is sent to the balanced phosphoric-acid concentration setting-out system 92 as an operating temperature of T degrees C of said phosphoric acid fuel cell 10.

[0052] The amount m of the generation water in which phosphoric-acid concentration is reduced in the balanced phosphoric-acid concentration setting-out system 92 (mol/min) Ratio a (%) which evaporates from the inside of the amount of the water in which phosphoric-acid concentration is reduced, and a fuel gas side is inputted, and when it is the operating temperature of T degrees C of a phosphoric acid fuel cell 10 So that the phosphoric-acid concentration to which the amount of the generation water in which phosphoric-acid concentration is reduced, and the amount of the water which evaporates out of a phosphoric acid are balancing may turn into more than the criteria phosphoric-acid concentration that the performance degradation of a proper does not produce in said phosphoric acid fuel cell 10 The fuel gas utilization factor r_a , the rate r_c of oxidizer gas utilization, the operating pressure P_a by the side of fuel gas, the operating pressure P_c by the side of oxidizer gas, fuel gas temperature T_a^{**} , and oxidizer gas-temperature T_c^{**} are set up if needed.

[0053] Here, the set-up fuel gas utilization factor r_a is sent to the quantity-of-gas-flow control system 84 by the side of fuel gas. A required fuel gas flow rate is set up from the fuel gas utilization factor r_a and the amount of fuel gas consumed by the reaction, and a fuel gas flow rate is controlled by this quantity-of-gas-flow control system 84 through the quantity-of-gas-flow controller 56. The set-up rate r_c of oxidizer gas utilization is similarly sent to quantity-of-gas-flow control-system 84a by the side of oxidizer gas. A required oxidizer quantity of gas flow is set up from the rate r_c of oxidizer gas utilization, and the oxidizer capacity consumed by the reaction, and an oxidizer quantity of gas flow is controlled by this quantity-of-gas-flow control-system 84a through quantity-of-gas-flow controller 56a.

[0054] Moreover, the operating pressure P_a by the side of the set-up fuel gas is sent to the pressure control system 90 by the side of fuel gas, and the input pressure or the output pressure of a phosphoric acid fuel cell 10 is controlled by the set point by adjusting a back-pressure valve 72. The operating pressure P_c by the side of the set-up oxidizer gas is similarly sent to pressure-control-system 90a by the side of oxidizer gas, and the input pressure or the output pressure of a phosphoric acid fuel cell 10 is controlled by the set point by adjusting back-pressure valve 72a.

[0055] Set-up fuel gas temperature T_a^{**} is sent to the gas-temperature control system 86 by the side of fuel gas, and controls fuel gas by the heating heater 62 to the set point further again. Oxidizer gas-temperature T_c^{**} set up similarly is sent to gas-temperature control-system 86a by the side of oxidizer gas, and oxidizer gas is controlled by heating heater 62a by the set point.

[0056] In addition, the ratio which evaporates from the inside of the amount of the water in which the operating conditions, i.e., the current density, the phosphoric acid fuel cell operating temperature, the fuel gas temperature, the oxidizer gas temperature, the fuel gas utilization factor, the rate of oxidizer gas utilization, and phosphoric-acid concentration in the minimum operating temperature which performance degradation does not produce are reduced, and a fuel gas side is made to memorize in a phosphoric acid fuel cell 10. And at the time of a start up, after carrying out temperature up to the minimum operating temperature which does not produce the above-mentioned performance degradation, operation is started based on the conditions the above was remembered to be.

[0057] Thus, when operating a phosphoric acid fuel cell 10 with this operation gestalt according to less than 120-degree C a temperature requirement and low-power output operational status, Even if the ratio in which produced water exists as liquefied water is high and it is the case where reduce the concentration of a phosphoric acid and the amount of phosphoric acids is made to increase Operating conditions are set up so that the phosphoric-acid concentration at the

time of the amount of the generation water in which phosphoric-acid concentration is reduced, and the amount of the water which evaporates out of a phosphoric acid balancing may become more than the criteria (that is, desired engine performance can be maintained) phosphoric-acid concentration which performance degradation does not produce. Therefore, additional equipment of a large-sized heater, the phosphoric-acid makeup equipment according to individual, etc. becomes unnecessary, generation-of-electrical-energy performance degradation and performance degradation are certainly prevented with an easy and economical configuration, and the effect that the good generation of electrical energy in the time of low temperature becomes executable is acquired.

[0058] [Example]

In the decision phosphoric acid fuel cell 10 of the phosphoric-acid concentration which the degradation of a proper does not produce in a phosphoric acid fuel cell 10, if a phosphoric acid flows out, the cell engine performance will not once return to the initial engine performance. For this reason, after operating by operating conditions to which generation water arises and phosphoric-acid concentration is reduced, the phosphoric-acid concentration at the time of the minimum of the operating temperature which can be returned to the initial engine performance turns into criteria phosphoric-acid concentration which the performance degradation of a proper does not produce in a phosphoric acid fuel cell 10.

[0059] Then, conditions without the performance degradation accompanying runoff of a phosphoric acid, i.e., the operating temperature of a phosphoric acid fuel cell 10, evaluated the engine performance (current density - voltage characteristic) of said phosphoric acid fuel cell 10 under the conditions 50% and whose rate of oxidizer gas utilization 201.3kPa(s) and a fuel gas utilization factor are [the operating pressure by the side of 160 degrees C and fuel gas] 50% for the operating pressure by the side of 201.3kPa(s) and oxidizer gas, and voltage in case current density is 1 A/cm² was made into the criteria of the initial engine performance.

[0060] Next, the operating temperature of a phosphoric acid fuel cell 10 was made into the operating temperature of T degrees C lower than 160 degrees C, and according to the same conditions as the above, with the fixed current density of 0.2 A/cm², other conditions continued for 3 hours and performed the generation of electrical energy. And it asked for the ratio a which evaporates from the inside of the amount of the water in which the amount of the water in which the concentration of a phosphoric acid is reduced reduces the concentration of the measured value of the dew-point sensors 67 and 67a to a phosphoric acid further, and a fuel gas side from current density and effective electrode area. Then, again, the operating temperature was made into 160 degrees C, the engine performance of a phosphoric acid fuel cell 10 was evaluated, and the voltage at current density 1 A/cm² was measured.

[0061] Subsequently, when asked for the operating temperature to which conditions with an operating temperature of T degrees C are changed into, and the voltage at current density 1 A/cm² before and behind a repeat deed and a low-temperature generation of electrical energy does not fall the above-mentioned measurement rather than the initial engine performance, it was 120 degrees C. The ratio a which evaporates from the inside of the amount of the water in which phosphoric-acid concentration is reduced, and a fuel gas side was 20%. And the inside of the amount of the water in which 0.2 A/cm² and the effective electrode area S reduce [operating temperature / of T degrees C / current density / I / 120 degrees C and] the concentration of 2 and a phosphoric acid by 196cm, The fuel gas utilization factor ra 20% 50%, [the ratio a which evaporates from a fuel gas side] 0 degree C whose rate rc of oxidizer gas utilization is reference condition, and the fuel gas flow rate fa converted into 1atm 50% 0.54 l/min, For 0 degree C which is reference condition, and the oxidizer quantity of gas flow fc converted into 1atm, the operating pressure Pa by the side of 1.33 l/min and fuel gas was [the operating pressure Pc by the side of 201.3kPa(s) and oxidizer gas] 201.3kPa(s).

[0062] When the amount of the generation water in which phosphoric-acid concentration is reduced, and the amount of the water which evaporates out of a phosphoric acid were balancing, the maximum vapor tension Ps and a of the phosphoric acid in fuel gas is 33.6kPa(s), and the maximum vapor tension Ps and c of the phosphoric acid in oxidizer gas was set to 31.8kPa(s) from these. At this time, the phosphoric-acid concentration by the side of fuel gas is 84.9%, and the phosphoric-acid concentration by the side of oxidizer gas became 86.6%. Therefore, the minimum of the phosphoric-acid concentration which does not produce the performance degradation of a proper in a phosphoric acid fuel cell 10 became 84.9%.

[0063] In the case of 80-degree C operating temperature, first, the fuel gas utilization factor ra, the rate rc of oxidizer gas utilization, the operating pressure Pa by the side of fuel gas, and operating pressure Pc by the side of oxidizer gas were usually made into the same conditions as the time of operation, and the degree of degradation was measured. Then, the operating pressure by the side of 201.3kPa(s) and oxidizer gas was set up as 201.3kPa(s), the rate of oxidizer gas utilization was set [the operating temperature of the phosphoric acid fuel cell 10 without performance degradation / the operating pressure by the side of 160 degrees C and fuel gas] up for the fuel gas utilization factor to 50% 50%, the engine performance (current density - voltage characteristic) of a phosphoric acid fuel cell 10 (an active electrode area is

2 196cm) was evaluated under these conditions (henceforth typical operating conditions), and voltage it will be whose current density in 1 A/cm² was made into the initial performance standard.

[0064] Then, the conditions of the operating pressure by the side of oxidizer gas (201.3kPa), a fuel gas utilization factor (50%), and the rate (50%) of oxidizer gas utilization were made the same, the operating temperature was reduced to 80 degrees C, generating current density on condition that 0.2 A/cm², in this condition, it continued for 3 hours and the generation of electrical energy was performed. In addition, the ratio which evaporates from the inside of the amount of the water in which the concentration of a phosphoric acid is reduced, and a fuel gas side was 50%.

[0065] When the engine performance of a phosphoric acid fuel cell 10 was evaluated under the typical operating conditions on the basis of the above-mentioned initial engine performance, the voltage it will be whose current density in 1 A/cm² was measured and this value was compared with the initial performance standard after the generation of electrical energy of 3 hours, that voltage retention is 78% and performance degradation had produced it, without the ability returning to the initial engine performance.

[0066] Next, the existence of the performance degradation at the time of usually reducing the fuel gas utilization factor r_a , the rate r_c of oxidizer gas utilization, the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas rather than the time of operation was verified. The inside of the amount of the water in which 80 degrees C and current density are reduced to in the operating temperature of a phosphoric acid fuel cell 10, and 0.2 A/cm² and an active electrode area specifically reduce 2 and early phosphoric-acid concentration by 196cm, So that the ratio which evaporates from a fuel gas side may be made into 50% and phosphoric-acid concentration may become 84.9% or more which is the minimum of the criteria phosphoric-acid concentration which does not produce performance degradation. The combination of the fuel gas utilization factor r_a , the rate r_c of oxidizer gas utilization, the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas was searched for.

[0067] In the phosphoric acid whose phosphoric-acid concentration is 84.9%, the maximum vapor tension in 80 degrees C is 6.10kPa(s), and set up the conditions which set maximum vapor tension of the phosphoric acid by the side of fuel gas and oxidizer gas to 6.10 or less kPas. That is, when the operating pressure P_a by the side of fuel gas and operating pressure P_c by the side of oxidizer gas were set to 101.3kPa(s) which are the minimum values (atmospheric pressure) of an output pressure, respectively, the rate r_c of oxidizer gas utilization was set up for the fuel gas utilization factor r_a to 10% 10% that what is necessary is just to make 11% or less and the rate r_c of oxidizer gas utilization 14% or less for the fuel gas utilization factor r_a .

[0068] Then, after obtaining an initial performance standard like the above based on typical operating conditions without performance degradation, the operating pressure P_a by the side of fuel gas and operating pressure P_c by the side of oxidizer gas are set to 101.3kPa(s) which are output pressures, respectively, 10%, 10% and current density generated electricity [the rate r_c of oxidizer gas utilization] on condition that 0.2 A/cm², and the fuel gas utilization factor r_a reduced the operating temperature to 80 degrees C.

[0069] After continuing for 3 hours and performing a generation of electrical energy, without changing operating conditions, the engine performance (current density - voltage characteristic) of a phosphoric acid fuel cell 10 was evaluated under the typical operating conditions which are the criteria of the initial engine performance, and the voltage it will be whose current density in 1 A/cm² was measured. When this value was compared with the initial performance standard, that voltage retention is 100% and performance degradation was not accepted.

[0070] In the case of 65-degree C operating temperature, the existence of the performance degradation at the time of usually reducing the fuel gas utilization factor r_a , the rate r_c of oxidizer gas utilization, the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas rather than the time of operation was verified. The ratio which evaporates from the inside of the amount of the water in which 65 degrees C and current density are reduced to in the operating temperature of a phosphoric acid fuel cell 10, and 0.2 A/cm² and an active electrode area reduce the concentration of 2 and an early phosphoric acid by 196cm, and a fuel gas side was made into 50%, and the combination of the fuel gas utilization factor r_a , the rate r_c of oxidizer gas utilization, the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas be searched for so that phosphoric-acid concentration might become 84.9% or more which is the minimum of the phosphoric-acid concentration which do produce performance degradation

[0071] In this case, in the phosphoric acid of 84.9% of phosphoric-acid concentration, the maximum vapor tension in 65 degrees C is 2.8kPa(s). Then, in order to set maximum vapor tension of the phosphoric acid by the side of fuel gas and oxidizer gas to 2.8 or less kPas, when reducing the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas to 101.3kPa(s) which are the minimum values (atmospheric pressure) of an output pressure, respectively, the fuel gas utilization factor r_a and the rate r_c of oxidizer gas utilization were made into 5%, respectively that what is necessary is just to make 5% or less and the rate r_c of oxidizer gas utilization 13% or less

for the fuel gas utilization factor r_a .

[0072] Next, the engine performance by typical operating conditions without performance degradation was evaluated, and voltage at current density 1 A/cm² was made into the initial performance standard. Then, the operating temperature was reduced to 65 degrees C, performing 5% for the rate r_c of oxidizer gas utilization, and generating electricity current density on condition that 0.2 A/cm² 5%, in 101.3kPa(s) and the fuel gas utilization factor r_a which are an output pressure about the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas. And after continuing for 3 hours and performing a generation of electrical energy, without changing operating conditions, the engine performance (current density - voltage characteristic) of a phosphoric acid fuel cell 10 was succeedingly evaluated under typical operating conditions, and the voltage it will be whose current density in 1 A/cm² was measured. When this value was compared with the initial performance standard, that voltage retention is 100% and performance degradation was not accepted.

[0073] In the case of 60-degree C operating temperature, the existence of the performance degradation at the time of usually reducing the fuel gas utilization factor r_a , the rate r_c of oxidizer gas utilization, the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas rather than the time of operation was verified. The ratio which evaporates from the inside of the amount of the water in which 60 degrees C and current density are reduced to in the operating temperature of a phosphoric acid fuel cell 10, and 0.2 A/cm² and an active electrode area reduce 2 and early phosphoric-acid concentration by 196cm, and a fuel gas side was made into 50%, and the combination of the fuel gas utilization factor r_a , the rate r_c of oxidizer gas utilization, the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas was searched for so that phosphoric-acid concentration might become 84.9% or more which is the minimum of the phosphoric-acid concentration which do produce performance degradation.

[0074] In the phosphoric acid of 84.9% of phosphoric-acid concentration, the maximum vapor tension in 60 degrees C serves as 2.2kPa(s). What is necessary is for the fuel gas utilization factor r_a just to reduce 4% or less and the rate r_c of oxidizer gas utilization to 10% or less, when reducing the operating pressure P_a by the side of fuel gas, and the operating pressure P_c by the side of oxidizer gas to 101.3kPa(s) which are the minimum values (atmospheric pressure) of an output pressure, respectively, in order to set maximum vapor tension of the phosphoric acid by the side of fuel gas and oxidizer gas to 2.2 or less kPas.

[0075] In the above-mentioned case, when the rate of gas utilization was not much low, it was not practical, and it was higher than the operating temperature in the temperature by the side of fuel gas and oxidizer gas, for example, was set as 120 degrees C, and verified whether performance degradation would arise. First, the operating temperature was reduced to 60 degrees C, generating [generate the rate r_c of oxidizer gas utilization, and / 101.3kPa(s) and the fuel gas utilization factor r_a which are an output pressure about the operating pressure by the side of fuel gas, and the operating pressure by the side of oxidizer gas / current density] 120 degrees C and oxidizer gas temperature for 0.2 A/cm² and fuel gas temperature on 120-degree C conditions 5% 5%, after obtaining an initial performance standard under typical operating conditions without performance degradation.

[0076] And after continuing for 3 hours and performing a generation of electrical energy, without changing operating conditions, succeedingly, the engine performance (current density - voltage characteristic) of a phosphoric acid fuel cell 10 was evaluated under typical operating conditions, and the voltage at current density 1 A/cm² was measured. When this value was compared with the initial performance standard, that voltage retention is 100% and performance degradation was not accepted.

[0077]

[Effect of the Invention] In the operating method of the phosphoric acid fuel cell concerning this invention The phosphoric-acid concentration at the time of the amount of said produced water to which phosphoric-acid concentration is reduced, and the amount of the water which evaporates out of a phosphoric acid balancing, in case it operates under the conditions in which produced water exists as liquefied water While avoiding generation-of-electrical-energy performance degradation certainly by setting up operating conditions so that it may become more than the criteria phosphoric-acid concentration which can maintain the desired engine performance, the initial engine performance is certainly maintainable at the time of a reboot. It will become economical, while additional equipment of a large-sized heater, phosphoric-acid makeup equipment, etc. becomes unnecessary and a configuration simplifies effectively by this.

[Translation done.]

* NOTICES *

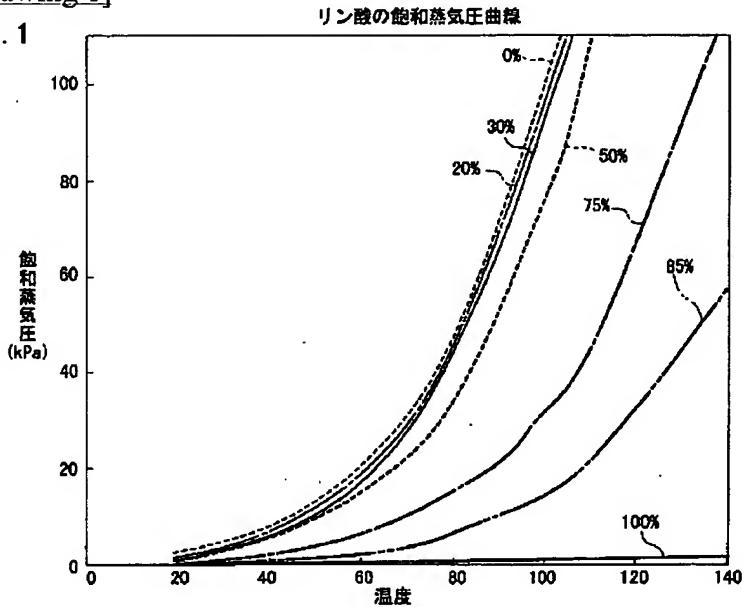
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DRAWINGS

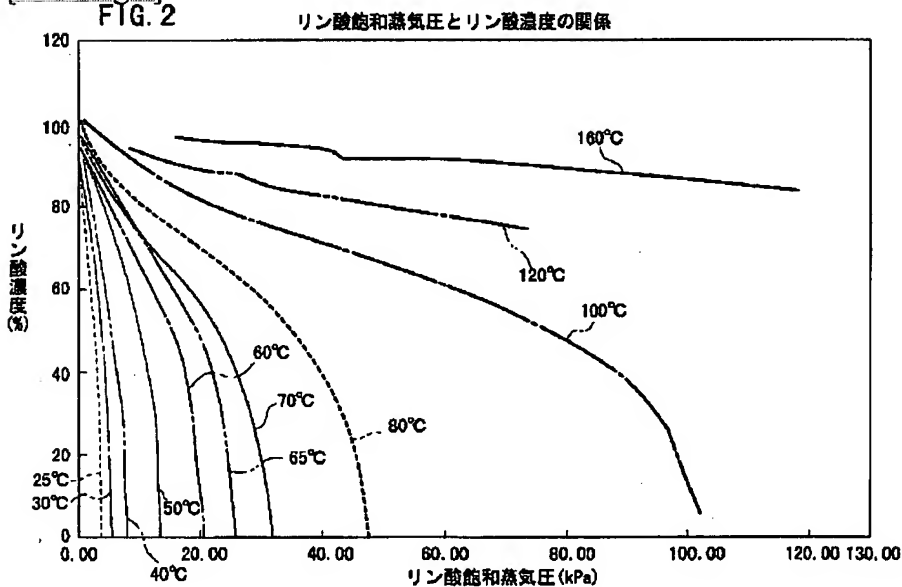
[Drawing 1]

FIG. 1



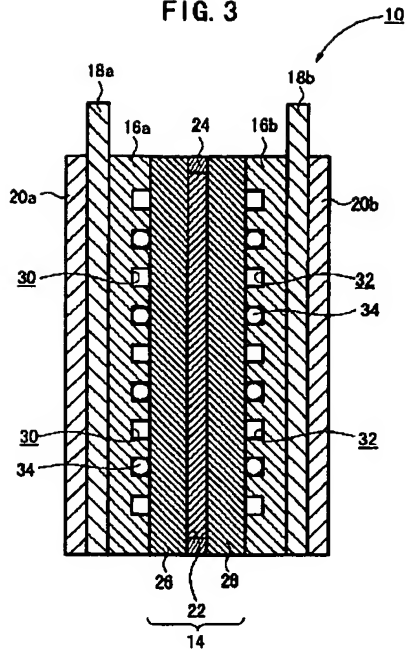
[Drawing 2]

FIG. 2



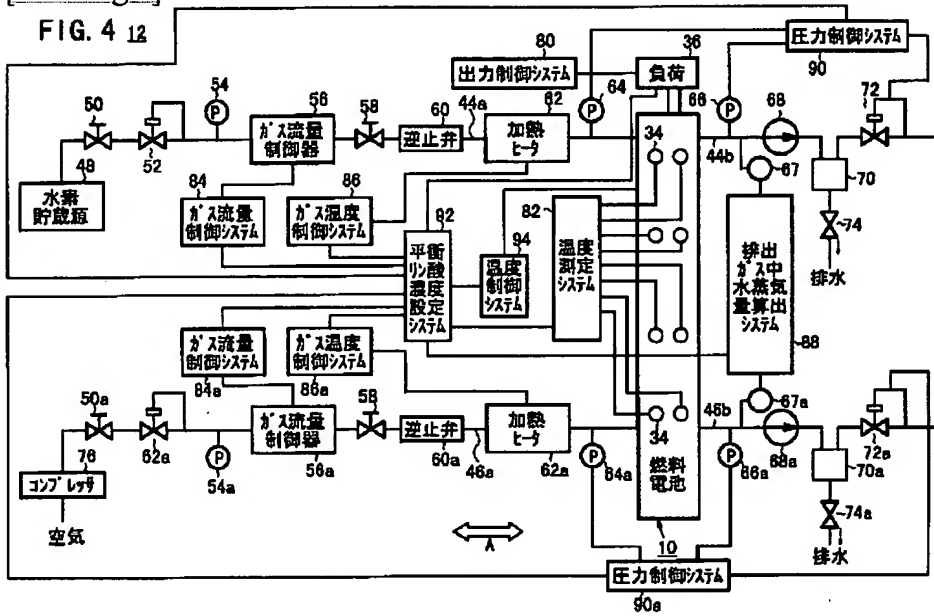
[Drawing 3]

FIG. 3



[Drawing 4]

FIG. 4 12



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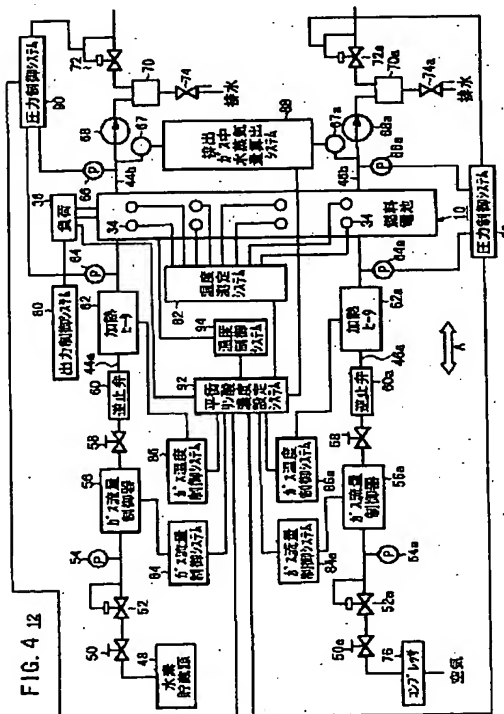
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(54)【発明の名称】リン酸型燃料電池の運転方法

(57)【要約】

【課題】追加設備を用いることなく、リン酸型燃料電池を低温下で良好に運転することを可能にする。

【解決手段】リン酸型燃料電池10を反応生成水が液状水として存在する条件下で運転する際に、リン酸濃度を低下させる前記反応生成水の量とリン酸中から蒸発する水の量とが平衡する際のリン酸濃度が、所望の性能を維持し得る基準リン酸濃度以上になるように運転条件を設定する。具体的には、平衡リン酸濃度設定システム92によりガス流量制御システム84、84a、ガス温度制御システム86、86aおよび圧力制御システム90、90aが必要に応じて制御され、リン酸型燃料電池10の性能劣化や発電性能の低下を有効に回避する。



【特許請求の範囲】

【請求項1】リン酸を含浸させた電解質をアノード側電極とカソード側電極とで挟んで構成される接合体を有し、燃料ガス流路を介して前記アノード側電極に燃料ガスを供給する一方、酸化剤ガス流路を介して前記カソード側電極に酸化剤ガスを供給するリン酸型燃料電池の運転方法であって、

反応生成水が液状水として存在する条件下で運転する際に、リン酸濃度を低下させる前記反応生成水の量とリン酸中から蒸発する水の量とが平衡する際のリン酸濃度が、所望の性能を維持し得る基準リン酸濃度以上になるように、運転条件を設定することを特徴とするリン酸型燃料電池の運転方法。

【請求項2】請求項1記載の運転方法において、燃料ガス利用率および酸化剤ガス利用率を設定することにより、前記リン酸濃度を前記基準リン酸濃度以上に制御することを特徴とするリン酸型燃料電池の運転方法。

【請求項3】請求項1または2記載の運転方法において、前記燃料ガス側の運転圧力および前記酸化剤ガス側の運転圧力を設定することにより、前記リン酸濃度を前記基準リン酸濃度以上に制御することを特徴とするリン酸型燃料電池の運転方法。

【請求項4】請求項1乃至3のいずれか1項に記載の運転方法において、前記燃料ガス側の温度および前記酸化剤ガス側の温度を運転温度よりも高温に設定することにより、前記リン酸濃度を前記基準リン酸濃度以上に制御することを特徴とするリン酸型燃料電池の運転方法。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、リン酸を含浸させた電解質をアノード側電極とカソード側電極とで挟んで構成される接合体を有し、燃料ガス供給系を介して前記アノード側電極に燃料ガスを供給する一方、酸化剤ガス供給系を介して前記カソード側電極に酸化剤ガスを供給するリン酸型燃料電池の運転方法に関する。

【0002】

【従来の技術】燃料電池の一形態であるリン酸型燃料電池(PAFC)は、例えば、液体電解質であるリン酸をポリベンズイミダゾール等の高分子膜に含浸させた電解質マトリックス層の両側に、それぞれカーボンを主体とするアノード側電極およびカソード側電極を対設して構成される電解質・電極接合体を、セパレータ(バイポーラ板)によって挟持することにより構成される発電セル(単位燃料電池セル)を備えており、通常、この発電セルを所定数だけ積層して燃料電池スタックとして使用している。

【0003】この種の燃料電池において、アノード側電極に供給された燃料ガス、例えば、主に水素を含有するガス(水素含有ガス)は、触媒電極上で水素がイオン化され、電解質を介してカソード側電極側へと移動する。

その間に生じた電子が外部回路に取り出され、直流の電気エネルギーとして利用される。なお、カソード側電極には、酸化剤ガス、例えば、主に酸素を含有するガスあるいは空気(酸素含有ガス)が供給されているために、このカソード側電極において、水素イオン、電子および酸素が反応して水が生成される。

【0004】上記のリン酸型燃料電池では、運転温度が比較的高温(120℃~190℃前後)に設定されており、一般的には、定常運転に至るまでの間、ヒータ等の加熱手段を使用して前記リン酸型燃料電池を加温する。そして、このリン酸型燃料電池が、略100℃以上まで昇温された後、前記リン酸型燃料電池を作動させることにより、発電に伴う自己発熱を利用して120℃~190℃まで昇温させている。

【0005】

【発明が解決しようとする課題】ところで、運転開始時のように、100℃未満の比較的低い温度下においては、リン酸型燃料電池の発電に伴う反応生成水が液状水として存在する比率が高くなるとともに、この液状水が水蒸気化し難くなっている。このため、液状水がリン酸の濃度を低下させてリン酸量を増加させ、アノード側電極およびカソード側電極に蓄積可能な量を超える液状水が存在してしまう。

【0006】このように、アノード側電極およびカソード側電極中にリン酸が溢れてしまうと、電解質マトリックス層中に保持されているリン酸が流出してしまい、この流出したリン酸が燃料ガス流路および酸化剤ガス流路を通過してリン酸型燃料電池本体の外部に排出されてしまう。これにより、リン酸型燃料電池の運転を停止した後、に再起動させた際、電池性能は初期性能を維持することができず、性能低下が惹起されるという問題が指摘されている。

【0007】さらに、アノード側電極中およびカソード側電極中にリン酸が溢れると、発電に必要な反応を促進させる機能を有する触媒の活性が低下し、あるいは燃料ガス流路や酸化剤ガス流路が閉塞されて反応ガス(燃料ガスおよび/または酸化剤ガス)が流通し難くなり、発電性能が低下するという問題がある。

【0008】そこで、リン酸の流出に伴う性能劣化を改善するために、個別のリン酸補給装置を用いることが考えられるが、燃料電池システム全体が大型化してしまうという問題がある。

【0009】また、リン酸型燃料電池を100℃以上まで迅速に昇温させるために、大型ヒータを用いることが考えられるが、同様に、燃料電池システム全体が大型化するとともに、経済的ではないという問題が指摘されている。

【0010】本発明はこの種の問題を解決するものであり、設備が大型化することがなく、簡単な構成および制御で、反応生成水による性能低下を確実に阻止すること

が可能なリン酸型燃料電池の運転方法を提供することを目的とする。

【0011】

【課題を解決するための手段】本発明に係るリン酸型燃料電池の運転方法では、反応生成水が液状水として存在する条件下で運転する際に、リン酸濃度を低下させる前記反応生成水の量とリン酸中から蒸発する水の量とが平衡する際のリン酸濃度が、所望の性能を維持し得る基準リン酸濃度以上になるように、運転条件を設定している。

【0012】通常、リン酸型燃料電池に用いられるリン酸は、5酸化2リンを水に溶解させたものであり、特に、リン酸型燃料電池に使用される高濃度のリン酸は吸湿性が高く、水と極めて混合し易い。この場合、リン酸の蒸気成分は、5酸化2リン（正確には2量体）と水であるが、5酸化2リンの蒸気圧は200℃付近まで非常に低くなっている。このため、リン酸の蒸気成分が略水であるものの、リン酸の蒸気圧は水の蒸気圧とは異なっている。

【0013】具体的には、リン酸濃度におけるリン酸の温度と飽和蒸気圧の関係は、図1の線図に示されており、図1中のリン酸濃度0%の線図が、水の飽和蒸気圧線図に相当している。これにより、リン酸の飽和蒸気圧はリン酸濃度により大きく変化しており、リン酸中の水はリン酸濃度が低いほど蒸発し易くなる一方、リン酸濃度が高いほど蒸発し難くなっている。

【0014】リン酸型燃料電池では、起動時のような低温時や低出力運転状態において、前記リン酸型燃料電池の温度が通常作動温度（120℃～190℃前後）よりも低下した際に、リン酸中の水の蒸発速度が遅くなり、蒸発する水の量よりも生成水の量が多い状態となる。従って、電解質・電極接合体中のリン酸濃度が低くなる一方、リン酸の飽和蒸気圧が高くなって水が蒸発し易くなる。

【0015】このため、生成水の量に応じてリン酸濃度が低下され、ある時点で、リン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡に達する。この状態では、リン酸量が増加しており、このリン酸量が電解質・電極接合体に蓄えられるリン酸量よりも多くなってしまうと、リン酸が燃料ガス流路および酸化剤ガス流路からリン酸型燃料電池の外部に排出されて性能劣化が生じるおそれがある。

【0016】これにより、リン酸濃度を低下させる反応生成水の量とリン酸中から蒸発する水の量とが平衡する際のリン酸濃度が、発電性能を含む性能の劣化が生じない基準リン酸濃度以上になるように運転条件を設定すれば、リン酸型燃料電池の温度が低い場合であっても性能劣化を惹起することがなく、効率的な発電が遂行可能に

なる。

【0017】また、性能劣化が生じないリン酸濃度は、電解質・電極接合体内の内部構成や寸法等に影響され、そのリン酸型燃料電池に固有のものである。従って、所望の性能を維持し得るリン酸濃度を、予め実験的に設定しておく必要がある。

【0018】運転条件の設定

電流密度が I (A/cm^2) で電極有効面積が S (cm^2) のとき、単位時間あたりに生成される水の量、すなわち、リン酸の濃度を低下させる水の量 m (mol/min) は、燃料ガス利用率 ra (%) および酸化剤ガス利用率 rc (%) に関係なく一定であり、下記の(1)式により求められる。

【0019】

【数1】

$$m = I \times S \times \frac{60}{96500 \times 2} \dots\dots(1)$$

【0020】次に、運転温度が T ℃、燃料ガス温度が T_a ℃、酸化剤ガス温度が T_c ℃、燃料ガス側運転圧力が P_a (kPa)、酸化剤側運転圧力が P_c (kPa)、燃料ガスの供給流量（燃料ガス流量）が f_a (normal l/min)（但し、normal l/min は標準状態である0℃、1atmに換算した流量である）、酸化剤ガスの供給流量（酸化剤ガス流量）が f_c (normal l/min)、燃料ガス側から排出されるガス流量が f_{ea} (normal l/min)、酸化剤ガス側から排出されるガス流量が f_{ec} (normal l/min)、リン酸の濃度を低下させる水の量の中、燃料ガス側から蒸発する比率を a (%)、および酸化剤ガス側から蒸発する比率を $1-a$ (%) とする。

【0021】燃料ガス側から排出されるガス流量 f_{ea} は、発電により消費されなかった燃料ガスの流量と、リン酸の濃度を低下させる水の量の中、燃料ガス側から蒸発する水蒸気量との和である（下記の(2)式より求められる）。

【0022】

【数2】

$$f_{ea} = f_a \times \frac{100-ra}{100} + m \times \frac{a}{100} \times 22.4 \dots\dots(2)$$

【0023】そこで、燃料ガス温度 T_a ℃と酸化剤ガス温度 T_c ℃が運転温度 T ℃と等しいとき、リン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡となった時点での排出燃料ガス中のリン酸の飽和蒸気圧 P_s 、 a は、下記の(3)式により求められる。

【0024】

【数3】

$$P_{s,a} = \frac{m \times \frac{a}{100} \times 22.4}{f_{ea}} \times P_a$$

$$= \frac{m \times \frac{a}{100} \times 22.4}{f_a \times \frac{100-ra}{100} + m \times \frac{a}{100} \times 22.4} \times P_a \quad \dots\dots(3)$$

【0025】一方、酸化剤ガス（空気であり、酸素が21wt%で、窒素が79wt%）側から排出されるガス流量 f_{ec} は、下記の（4）式より求められる。

$$f_{ec} = f_c \times (0.21 \times \frac{100-rc}{100} + 0.79) + m \times \frac{100-a}{100} \times 22.4 \quad \dots\dots(4)$$

【0027】さらに、排出酸化剤ガス中のリン酸の飽和蒸気圧 $P_{s,c}$ は同様に、下記の（5）式から求められる。

$$P_{s,c} = \frac{m \times \frac{100-a}{100} \times 22.4}{f_{ec}} \times P_c$$

$$= \frac{m \times \frac{100-a}{100} \times 22.4}{f_c \times (0.21 \times \frac{100-rc}{100} + 0.79) + m \times \frac{100-a}{100} \times 22.4} \times P_c \quad \dots\dots(5)$$

【0029】また、リン酸の飽和蒸気圧とリン酸濃度の関係は、図2の線図に示されており、温度 $T^{\circ}\text{C}$ におけるリン酸濃度は、リン酸の飽和蒸気圧の関数として求められる。図2から読解されるように、一定の運転温度 $T^{\circ}\text{C}$ において、リン酸の飽和蒸気圧が低いほどリン酸濃度は高くなっている。従って、排出燃料ガス中のリン酸の飽和蒸気圧 $P_{s,a}$ および酸化剤ガス中のリン酸の飽和蒸気圧 $P_{s,c}$ を用い、運転温度 $T^{\circ}\text{C}$ においてリン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡となるリン酸濃度を、それぞれ求めることができる。

【0030】具体的には、排出燃料ガス中のリン酸の飽和蒸気圧 $P_{s,a}$ は、燃料ガス側の運転圧力 P_a 、燃料ガス側から排出されるガス流量 f_{ea} の関数である。上記の排出燃料ガス流量 f_{ea} は、リン酸の濃度を低下させる水の量 m および燃料ガス利用率 r_a の関数であり、 m が一定のときに燃料ガス利用率 r_a が低いほど前記排出燃料ガス流量 f_{ea} が多くなる。従って、排出燃料ガス中のリン酸の飽和蒸気圧 $P_{s,a}$ は、燃料ガス側の運転圧力 P_a が低いとき、または燃料ガス利用率 r_a が低いときに低くなり、この結果、リン酸濃度が高くなる。

【0031】同様に、排出酸化剤ガス中のリン酸の飽和蒸気圧 $P_{s,c}$ は、酸化剤ガス側の運転圧力 P_c 、酸化剤ガス利用率 r_c および標準状態である 0°C 、 1atm に換算した酸化剤ガス流量 f_c の関数である。従って、排出酸化剤ガス中のリン酸の飽和蒸気圧 $P_{s,c}$ は、酸化剤ガス側の運転圧力 P_c が低いとき、または酸化剤ガス利用率 r_c が低いときに低くなり、この結果、リン酸濃度が高くなる。

【0032】これにより、燃料ガス利用率 r_a と酸化剤

【0026】
【数4】

【0028】
【数5】

ガス利用率 r_c および/または燃料ガス側の運転圧力 P_a と酸化剤ガス側の運転圧力 P_c を設定することにより、リン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡となるリン酸濃度が、性能劣化を起こさない、すなわち、所望の性能を維持し得る基準リン酸濃度以上となり、低温運転時でも性能低下を確実に阻止することができる。

【0033】一方、リン酸濃度は、リン酸の飽和蒸気圧と温度との関数として求められる（図2参照）。このため、リン酸の飽和蒸気圧が一定の場合、リン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡となる温度を高く設定すれば、平衡時のリン酸濃度を高くすることが可能になる。

【0034】すなわち、燃料ガス温度 $T_a^{\circ}\text{C}$ および酸化剤ガス温度 $T_c^{\circ}\text{C}$ を運転温度 $T^{\circ}\text{C}$ よりも高くすることにより、リン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡となる際の平衡界面でのリン酸温度 $T'^{\circ}\text{C}$ を運転温度 $T^{\circ}\text{C}$ よりも高くし、平衡時のリン酸濃度を高くすることができる。なお、燃料ガス温度 $T_a^{\circ}\text{C}$ および酸化剤ガス温度 $T_c^{\circ}\text{C}$ と平衡界面でのリン酸温度 $T'^{\circ}\text{C}$ の関係は、そのリン酸型燃料電池に固有のものであり、実験的に求めておくことが望ましい。

【0035】

【発明の実施の形態】図3は、本発明の実施形態に係る運転方法を実施するためのリン酸型燃料電池10の構成説明図であり、図4は、前記リン酸型燃料電池10が組み込まれる燃料電池システム12の概略構成図である。

【0036】リン酸型燃料電池10は、図3に示すように、電解質・電極接合体14と、この電解質・電極接合体14の両面に配置されるバイポーラ板であるセパレー

タ 16 a、16 b と、前記セパレータ 16 a、16 b の外側に配置される集電用電極 18 a、18 b とを備える。集電用電極 18 a、18 b の外側には、エンドプレート 20 a、20 b が配置され、前記エンドプレート 20 a、20 b が図示しないボルトにより締め付けられることにより、リン酸型燃料電池 10 が一体的に構成される。

【0037】電解質・電極接合体 14 は、塩基性ポリマー、例えば、ポリベンズイミダゾール膜等の高分子膜にリン酸を含浸させた電解質マトリックス層 22 と、前記電解質マトリックス層 22 の両面に額縁状スペーサ 24 を介して対設されるアノード側電極 26 およびカソード側電極 28 とを備える。

【0038】セパレータ 16 a、16 b には、アノード側電極 26 およびカソード側電極 28 に対向する面に燃料ガス流路 30 と酸化剤ガス流路 32 とが形成されている。セパレータ 16 a、16 b には、アノード側電極 26 とカソード側電極 28 との境界面に対応して複数の温度センサ 34 が配置されている。

【0039】図 4 に示すように、リン酸型燃料電池 10 を構成する集電用電極 18 a、18 b には、モータ等の負荷 36 が接続されている。リン酸型燃料電池 10 には、燃料ガス流路 30 に連通する燃料ガス供給経路 44 a および燃料ガス排出経路 44 b と、酸化剤ガス流路 32 に連通する酸化剤ガス供給経路 46 a および酸化剤ガス排出経路 46 b とが接続される。

【0040】燃料ガス供給経路 44 a には、上流側からリン酸型燃料電池 10 の入口側に向かって燃料ガスである水素含有ガスを高圧で供給するための水素貯蔵源 48 と、電磁弁 50 と、減圧弁 52 と、圧力センサ 54 と、ガス流量制御器 56 と、遮断弁 58 と、逆止弁 60 と、加熱ヒータ 62 と、入口側圧力センサ 64 とが配置されている。燃料ガス排出経路 44 b には、出口側圧力センサ 66 と、露点センサ 67 と、熱交換器 68 と、気液分離器 70 と、背圧弁 72 とが配置されるとともに、この気液分離器 70 の出口側に電磁弁 74 が設けられている。

【0041】酸化剤ガス供給経路 46 a および酸化剤ガス排出経路 46 b は、上記の燃料ガス供給経路 44 a および燃料ガス排出経路 44 b と同様に構成されており、同一の構成要素には同一の参照数字に符号 a を付して、その詳細な説明は省略する。この酸化剤ガス供給経路 46 a の上流側には、酸化剤ガスとして空気を供給するためのコンプレッサ 76 が配置されている。

【0042】負荷 36 には、出力制御システム 80 が接続されるとともに、リン酸型燃料電池 10 に組み込まれている複数の温度センサ 34 が温度測定システム 82 に接続される。ガス流量制御器 56、56 a にガス流量制御システム 84、84 a が接続されるとともに、加熱ヒータ 62、62 a にガス温度制御システム 86、86 a

が接続される。露点センサ 67、67 a に排出ガス中水蒸気量算出システム 88 が接続され、入口側圧力センサ 64、64 a、出口側圧力センサ 66、66 a および背圧弁 72、72 a に圧力制御システム 90、90 a が接続される。

【0043】温度測定システム 82 は平衡リン酸濃度設定システム 92 に接続されるとともに、この平衡リン酸濃度設定システム 92 には、負荷 36、ガス流量制御システム 84、84 a、ガス温度制御システム 86、86 a、排出ガス中水蒸気量算出システム 88、圧力制御システム 90、90 a および温度制御システム 94 が接続される。

【0044】このように構成されるリン酸型燃料電池 10 を組み込む燃料電池システム 12 の動作について、以下に説明する。

【0045】燃料ガス供給経路 44 a 側では、水素貯蔵源 48 から高圧の水素含有ガスが供給され、減圧弁 52 を介して運転圧力の最大値の圧力に減圧された後、ガス流量制御器 56 によりガス利用率が制御される。この水素含有ガスは、加熱ヒータ 62 により所定温度まで昇温された後、背圧弁 72 が制御されることによって所定の運転圧力に調整され、リン酸型燃料電池 10 を構成するアノード側電極 26 側のセパレータ 16 a に設けられている燃料ガス流路 30 に導入される。

【0046】燃料ガス流路 30 に供給された水素含有ガスは、ガス利用率で規定される量の水素ガスをアノード側電極 26 中に拡散し、触媒層内で触媒反応により消費されて発電に寄与する。水素含有ガスの残りの部分は、リン酸濃度を低下させる生成水の中、燃料ガス側から排出される量を含んで燃料ガス排出経路 44 b 側に排出される。

【0047】一方、酸化剤ガス供給経路 46 a 側では、酸素含有ガス、例えば、空気がコンプレッサ 76 を介して高圧化された後、減圧弁 52 a により運転圧力の最大値の圧力に減圧される。この空気は、ガス流量制御器 56 a を通ってガス利用率が制御された後、加熱ヒータ 62 a で設定温度まで昇温される。

【0048】さらに、空気は背圧弁 72 a が制御されることによって所定の運転圧力に調整され、リン酸型燃料電池 10 のカソード側電極 28 側のセパレータ 16 b に形成されている酸化剤ガス流路 32 に導入される。酸化剤ガス流路 32 に供給された空気は、ガス利用率で規定される量がカソード側電極 28 中に拡散して触媒層内で触媒反応により消費され、発電に寄与する。この空気の残りの部分は、リン酸濃度を低下させる生成水の中、酸化剤ガス側から排出される量を含んで酸化剤ガス流路 32 を通って酸化剤ガス排出経路 46 b に排出される。

【0049】この場合、出力制御システム 80 により、その時点での要求出力から必要な電流値が設定され、この電流値に基づいてリン酸型燃料電池 10 と負荷 36 と

からなる閉回路に流す電子量が決定される。そして、閉回路に流す電子量に応じて消費される燃料ガスおよび酸化剤ガスの量が決定され、リン酸濃度を低下させる生成水の量 m (mol/min) が決定される。

【0050】次に、排出される燃料ガスおよび酸化剤ガスの露点が、リン酸型燃料電池10の出口側に接続された露点センサ67、67aを介して検出される。この露点データは、排出ガス中水蒸気量算出システム88に送られ、この排出ガス中水蒸気量算出システム88において、燃料ガス側に排出されるリン酸からの水分および酸化剤ガス側に排出されるリン酸からの水分が算出される。さらに、リン酸の濃度を低下させる水の量の中、燃料ガス側から蒸発する比率 a (%) が算出され、この比率 a (%) が平衡リン酸濃度設定システム92に送られる。

【0051】一方、リン酸型燃料電池10の運転温度は、このリン酸型燃料電池10内に配置された複数の温度センサ34を介して温度測定システム82により検出され、この検出された温度の最低値が、前記リン酸型燃料電池10の運転温度 $T^{\circ}\text{C}$ として平衡リン酸濃度設定システム92に送られる。

【0052】平衡リン酸濃度設定システム92では、リン酸濃度を低下させる生成水の量 m (mol/min)、リン酸濃度を低下させる水の量の中、燃料ガス側から蒸発する比率 a (%) が入力されており、リン酸型燃料電池10の運転温度 $T^{\circ}\text{C}$ のときに、リン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡となるリン酸濃度が、前記リン酸型燃料電池10に固有の性能劣化が生じない基準リン酸濃度以上となるように、燃料ガス利用率 r_a 、酸化剤ガス利用率 r_c 、燃料ガス側の運転圧力 P_a 、酸化剤ガス側の運転圧力 P_c 、燃料ガス温度 $T_a^{\circ}\text{C}$ および酸化剤ガス温度 $T_c^{\circ}\text{C}$ が必要に応じて設定される。

【0053】ここで、設定された燃料ガス利用率 r_a は、燃料ガス側のガス流量制御システム84に送られる。このガス流量制御システム84では、燃料ガス利用率 r_a と、反応により消費される燃料ガス量とから必要な燃料ガス流量を設定し、ガス流量制御器56を介して燃料ガス流量が制御される。設定された酸化剤ガス利用率 r_c は、同様に酸化剤ガス側のガス流量制御システム84aに送られる。このガス流量制御システム84aでは、酸化剤ガス利用率 r_c と反応により消費される酸化剤ガス量とから必要な酸化剤ガス流量が設定され、ガス流量制御器56aを介して酸化剤ガス流量が制御される。

【0054】また、設定された燃料ガス側の運転圧力 P_a は、燃料ガス側の圧力制御システム90に送られ、背圧弁72を調整することによってリン酸型燃料電池10の入口側圧力または出口側圧力が設定値に制御される。設定された酸化剤ガス側の運転圧力 P_c は、同様に酸化

剤ガス側の圧力制御システム90aに送られ、背圧弁72aが調整されることによって、リン酸型燃料電池10の入口側圧力または出口側圧力が設定値に制御される。

【0055】さらにまた、設定された燃料ガス温度 $T_a^{\circ}\text{C}$ は、燃料ガス側のガス温度制御システム86に送られ、加熱ヒータ62により燃料ガスを設定値に制御する。同様に設定された酸化剤ガス温度 $T_c^{\circ}\text{C}$ は、酸化剤ガス側のガス温度制御システム86aに送られ、加熱ヒータ62aにより酸化剤ガスが設定値に制御される。

【0056】なお、リン酸型燃料電池10において、性能劣化が生じない最低運転温度での運転条件、すなわち、電流密度、リン酸型燃料電池運転温度、燃料ガス温度、酸化剤ガス温度、燃料ガス利用率、酸化剤ガス利用率およびリン酸濃度を低下させる水の量の中、燃料ガス側から蒸発する比率を記憶させておく。そして、運転開始時には、上記の性能劣化を生じない最低運転温度まで昇温させた後、上記の記憶された条件に基づいて運転が開始される。

【0057】このように、本実施形態では、リン酸型燃料電池10を 120°C 未満の温度範囲や低出力運転状態で作動させる際、反応生成水が液状水として存在する比率が高く、リン酸の濃度を低下させてリン酸量を増加させる場合であっても、リン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡する際のリン酸濃度が、性能劣化が生じない（すなわち、所望の性能を維持し得る）基準リン酸濃度以上になるように運転条件が設定されている。従って、大型のヒータや個別のリン酸補給装置等の追加設備が不要となり、簡単かつ経済的な構成で、発電性能の低下や性能劣化を確実に阻止して、低温時での良好な発電が遂行可能になるという効果が得られる。

【0058】〔実施例〕

リン酸型燃料電池10に固有の性能低下が生じないリン酸濃度の決定

リン酸型燃料電池10では、一旦、リン酸が流出してしまうと、電池性能は初期性能に戻らない。このため、生成水が生じてリン酸濃度を低下させるような運転条件で運転した後に、初期性能に戻ることができる運転温度の下限時のリン酸濃度が、リン酸型燃料電池10に固有の性能劣化が生じない基準リン酸濃度となる。

【0059】そこで、リン酸の流出に伴う性能劣化がない条件、すなわち、リン酸型燃料電池10の運転温度が 160°C 、燃料ガス側の運転圧力が 201.3 kPa 、酸化剤ガス側の運転圧力が 201.3 kPa 、燃料ガス利用率が50%および酸化剤ガス利用率が50%の条件下で、前記リン酸型燃料電池10の性能（電流密度～電圧特性）を評価し、電流密度が $1\text{ A}/\text{cm}^2$ のときの電圧を初期性能の基準とした。

【0060】次に、リン酸型燃料電池10の運転温度を 160°C よりも低い運転温度 $T^{\circ}\text{C}$ とし、他の条件は上記

と同一の条件により、 0.2 A/cm^2 の一定電流密度で発電を3時間継続して行った。そして、電流密度と有効電極面積からリン酸の濃度を低下させる水の量を、さらに露点センサ67、67aの測定値からリン酸の濃度を低下させる水の量の中、燃料ガス側から蒸発する比率aを求めた。その後、再度、運転温度を 160°C とし、リン酸型燃料電池10の性能を評価して電流密度 1 A/cm^2 時の電圧を測定した。

【0061】次いで、運転温度 $T^\circ\text{C}$ の条件を変えて上記の測定を繰り返し行い、低温発電前後の電流密度 1 A/cm^2 時の電圧が初期性能よりも低下しない運転温度を求めたところ、 120°C であった。リン酸濃度を低下させる水の量の中、燃料ガス側から蒸発する比率aは20%であった。そして、運転温度 $T^\circ\text{C}$ が 120°C 、電流密度Iが 0.2 A/cm^2 、有効電極面積Sが 196 cm^2 、リン酸の濃度を低下させる水の量の中、燃料ガス側から蒸発する比率aが20%、燃料ガス利用率raが50%、酸化剤ガス利用率rcが50%、標準状態である 0°C 、 1 atm に換算した燃料ガス流量faが 0.54 l/min 、標準状態である 0°C 、 1 atm に換算した酸化剤ガス流量fcが 1.33 l/min 、燃料ガス側の運転圧力Paが 201.3 kPa および酸化剤ガス側の運転圧力Pcが 201.3 kPa であった。

【0062】これらから、リン酸濃度を低下させる生成水の量とリン酸中から蒸発する水の量とが平衡となった時点で、燃料ガス中のリン酸の飽和蒸気圧 $P_{s,a}$ が 33.6 kPa であり、酸化剤ガス中のリン酸の飽和蒸気圧 $P_{s,c}$ が 31.8 kPa となった。このとき、燃料ガス側のリン酸濃度は84.9%であり、酸化剤ガス側のリン酸濃度は86.6%となった。従って、リン酸型燃料電池10に固有の性能劣化を生じないリン酸濃度の下限は、84.9%となった。

【0063】運転温度 80°C の場合

まず、燃料ガス利用率ra、酸化剤ガス利用率rc、燃料ガス側の運転圧力Pa、酸化剤ガス側の運転圧力Pcを通常運転時と同一条件にし、性能低下の度合いを測定した。そこで、性能劣化のないリン酸型燃料電池10の運転温度を 160°C 、燃料ガス側の運転圧力を 201.3 kPa 、酸化剤ガス側の運転圧力を 201.3 kPa 、燃料ガス利用率を50%、酸化剤ガス利用率を50%に設定し、この条件（以下、基準運転条件という）下で、リン酸型燃料電池10（電極有効面積が 196 cm^2 ）の性能（電流密度～電圧特性）を評価し、電流密度が 1 A/cm^2 時の電圧を初期性能基準値とした。

【0064】続いて、酸化剤ガス側の運転圧力（ 201.3 kPa ）、燃料ガス利用率（50%）および酸化剤ガス利用率（50%）の条件を同一にし、電流密度を 0.2 A/cm^2 の条件で発電しつつ運転温度を 80°C まで低下させ、この状態で、発電を3時間継続して行った。なお、リン酸の濃度を低下させる水の量の中、燃料

ガス側から蒸発する比率は50%であった。

【0065】3時間の発電後に、上記の初期性能を基準とした基準運転条件下でリン酸型燃料電池10の性能を評価し、電流密度が 1 A/cm^2 時の電圧を測定し、この値を初期性能基準値と比較したところ、その電圧保持率は78%であり、初期性能に復帰できずに性能劣化が生じていた。

【0066】次に、燃料ガス利用率ra、酸化剤ガス利用率rc、燃料ガス側の運転圧力Paおよび酸化剤ガス側の運転圧力Pcを、通常運転時よりも低下させた場合の性能劣化の有無を検証した。具体的には、リン酸型燃料電池10の運転温度を 80°C 、電流密度を 0.2 A/cm^2 、電極有効面積を 196 cm^2 および初期のリン酸濃度を低下させる水の量の中、燃料ガス側から蒸発する比率を50%とし、リン酸濃度が性能劣化を生じない基準リン酸濃度の下限である84.9%以上となるように、燃料ガス利用率ra、酸化剤ガス利用率rc、燃料ガス側の運転圧力Paおよび酸化剤ガス側の運転圧力Pcの組み合わせを求めた。

【0067】リン酸濃度が84.9%のリン酸において、 80°C における飽和蒸気圧は 6.10 kPa であり、燃料ガス側および酸化剤ガス側のリン酸の飽和蒸気圧を 6.10 kPa 以下とする条件を設定した。すなわち、燃料ガス側の運転圧力Paおよび酸化剤ガス側の運転圧力Pcをそれぞれ出口側圧力の最低値（大気圧）である 101.3 kPa とすると、燃料ガス利用率raを11%以下、および酸化剤ガス利用率rcを14%以下にすればよく、例えば、燃料ガス利用率raを10%、酸化剤ガス利用率rcを10%に設定した。

【0068】そこで、上記と同様に、性能劣化のない基準運転条件に基づいて、初期性能基準値を得た後、燃料ガス側の運転圧力Paおよび酸化剤ガス側の運転圧力Pcをそれぞれ出口側圧力である 101.3 kPa とし、燃料ガス利用率raが10%、酸化剤ガス利用率rcが10%および電流密度が 0.2 A/cm^2 の条件で発電を行い、運転温度を 80°C まで低下させた。

【0069】運転条件を変えずに発電を3時間継続して行った後、初期性能の基準である基準運転条件下でリン酸型燃料電池10の性能（電流密度～電圧特性）を評価し、電流密度が 1 A/cm^2 時の電圧を測定した。この値を初期性能基準値と比較したところ、その電圧保持率は100%であって、性能劣化は認められなかった。

【0070】運転温度 65°C の場合

燃料ガス利用率ra、酸化剤ガス利用率rc、燃料ガス側の運転圧力Paおよび酸化剤ガス側の運転圧力Pcを、通常運転時よりも低下させた場合の性能劣化の有無を検証した。リン酸型燃料電池10の運転温度を 65°C 、電流密度を 0.2 A/cm^2 、電極有効面積を 196 cm^2 、および初期のリン酸の濃度を低下させる水の量の中、燃料ガス側から蒸発する比率を50%とし、リ

ン酸濃度が性能劣化を生じないリン酸濃度の下限である 84.9%以上となるように、燃料ガス利用率 r_a 、酸化剤ガス利用率 r_c 、燃料ガス側の運転圧力 P_a および酸化剤ガス側の運転圧力 P_c の組み合わせを求めた。

【0071】この場合、リン酸濃度 84.9%のリン酸において、65℃での飽和蒸気圧は 2.8 kPa である。そこで、燃料ガス側および酸化剤ガス側のリン酸の飽和蒸気圧を 2.8 kPa 以下とするためには、燃料ガス側の運転圧力 P_a および酸化剤ガス側の運転圧力 P_c をそれぞれ出口側圧力の最低値（大気圧）である 101.3 kPa まで低下させた場合に、燃料ガス利用率 r_a を 5%以下および酸化剤ガス利用率 r_c を 13%以下にすればよく、例えば、燃料ガス利用率 r_a および酸化剤ガス利用率 r_c をそれぞれ 5%とした。

【0072】次に、性能劣化のない基準運転条件による性能を評価し、電流密度 1 A/cm² 時の電圧を初期性能基準値とした。続いて、燃料ガス側の運転圧力 P_a および酸化剤ガス側の運転圧力 P_c を出口側圧力である 101.3 kPa、燃料ガス利用率 r_a を 5%、酸化剤ガス利用率 r_c を 5%、および電流密度を 0.2 A/cm² の条件で発電を行いながら、運転温度を 65℃まで低下させた。そして、運転条件を変えずに、発電を 3 時間継続して行った後、引き続き基準運転条件下でリン酸型燃料電池 10 の性能（電流密度～電圧特性）を評価し、電流密度が 1 A/cm² 時の電圧を測定した。この値を初期性能基準値と比較したところ、その電圧保持率は 100%であって、性能劣化は認められなかった。

【0073】運転温度 60℃の場合

燃料ガス利用率 r_a 、酸化剤ガス利用率 r_c 、燃料ガス側の運転圧力 P_a および酸化剤ガス側の運転圧力 P_c を、通常運転時よりも低下させた場合の性能劣化の有無を検証した。リン酸型燃料電池 10 の運転温度を 60℃、電流密度を 0.2 A/cm²、電極有効面積を 196 cm² および初期のリン酸濃度を低下させる水の量の中、燃料ガス側から蒸発する比率を 50%とし、リン酸濃度が性能劣化を生じないリン酸濃度の下限である 84.9%以上となるように、燃料ガス利用率 r_a 、酸化剤ガス利用率 r_c 、燃料ガス側の運転圧力 P_a および酸化剤ガス側の運転圧力 P_c の組み合わせを求めた。

【0074】リン酸濃度 84.9%のリン酸において、60℃での飽和蒸気圧は 2.2 kPa となる。燃料ガス側および酸化剤ガス側のリン酸の飽和蒸気圧を 2.2 kPa 以下とするためには、燃料ガス側の運転圧力 P_a および酸化剤ガス側の運転圧力 P_c をそれぞれ出口側圧力の最低値（大気圧）である 101.3 kPa まで低下させた場合、燃料ガス利用率 r_a を 4%以下および酸化剤ガス利用率 r_c を 10%以下に低下させればよい。

【0075】上記の場合、ガス利用率があまり低いと実用的ではなく、燃料ガスおよび酸化剤ガス側の温度を運転温度よりも高く、例えば、120℃に設定し、性能劣

化が生じないかを検証した。まず、性能劣化のない基準運転条件下で初期性能基準値を得た後、燃料ガス側の運転圧力および酸化剤ガス側の運転圧力を出口側圧力である 101.3 kPa、燃料ガス利用率 r_a を 5%、酸化剤ガス利用率 r_c を 5%、電流密度を 0.2 A/cm²、燃料ガス温度を 120℃および酸化剤ガス温度を 120℃の条件で発電しながら、運転温度を 60℃まで低下させた。

【0076】そして、運転条件を変えずに、発電を 3 時間継続して行った後、引き続き、基準運転条件下でリン酸型燃料電池 10 の性能（電流密度～電圧特性）を評価し、電流密度 1 A/cm² 時の電圧を測定した。この値を初期性能基準値と比較したところ、その電圧保持率は 100%であり、性能劣化は認められなかった。

【0077】

【発明の効果】本発明に係るリン酸型燃料電池の運転方法では、反応生成水が液状水として存在する条件下で運転する際に、リン酸濃度を低下させる前記反応生成水の量とリン酸中から蒸発する水の量とが平衡する際のリン酸濃度が、所望の性能を維持し得る基準リン酸濃度以上になるように運転条件を設定することにより、発電性能の低下を確実に回避するとともに、再起動時に初期性能を確実に維持することができる。これにより、大型ヒータやリン酸補給装置等の追加設備が不要になり、構成が有効に簡素化するとともに、経済的なものとなる。

【図面の簡単な説明】

【図 1】各リン酸濃度におけるリン酸の温度と飽和蒸気圧の関係を示す線図である。

【図 2】リン酸の飽和蒸気圧とリン酸濃度の関係を示す線図である。

【図 3】本発明に係る運転方法を実施するためのリン酸型燃料電池の構成説明図である。

【図 4】前記リン酸型燃料電池が組み込まれる燃料電池システムの概略構成図である。

【符号の説明】

10…リン酸型燃料電池	12…燃料電池システム
14…電解質・電極接合体	16a、16b…セパレータ
18a、18b…集電用電極	30…燃料ガス流路
32…酸化剤ガス流路	34…温度センサ
36…負荷供給経路	44a…燃料ガス供給経路
44b…燃料ガス排出経路	46a…酸化剤ガス供給経路
46b…酸化剤ガス排出経路	80…出力制御システム
82…温度測定システム	84、84a…ガス流量制御システム

86、86a...ガス温度制御システム

濃度設定システム

88...排出ガス中水蒸気量算出システム

94...温度制御システム

90、90a...圧力制御システム 92...平衡リン酸

【図1】

【図3】

FIG. 1

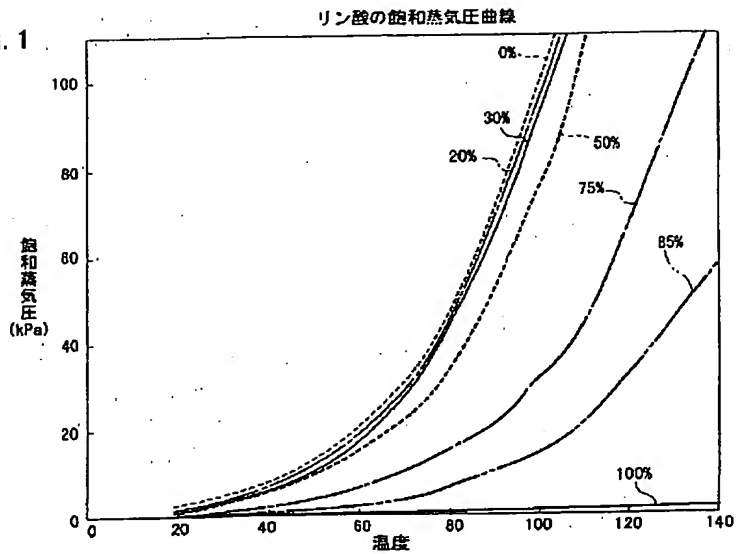
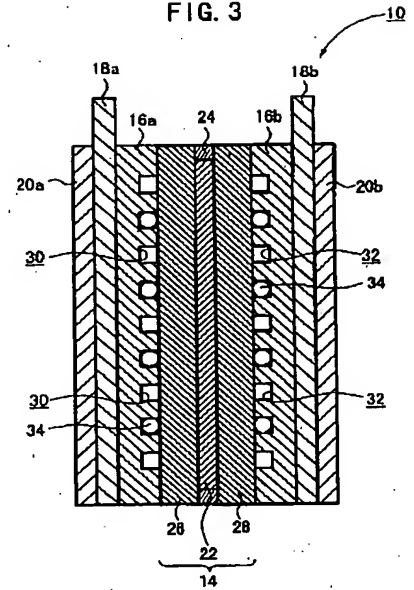
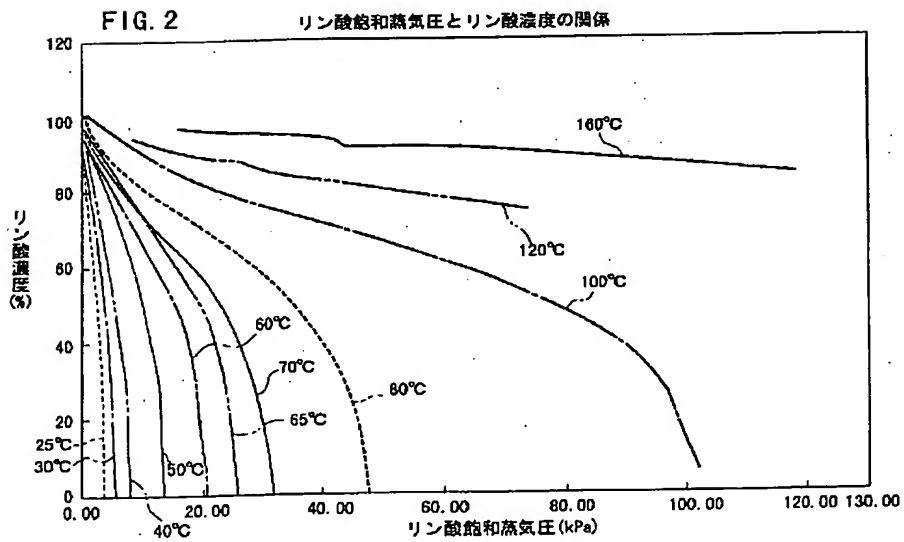


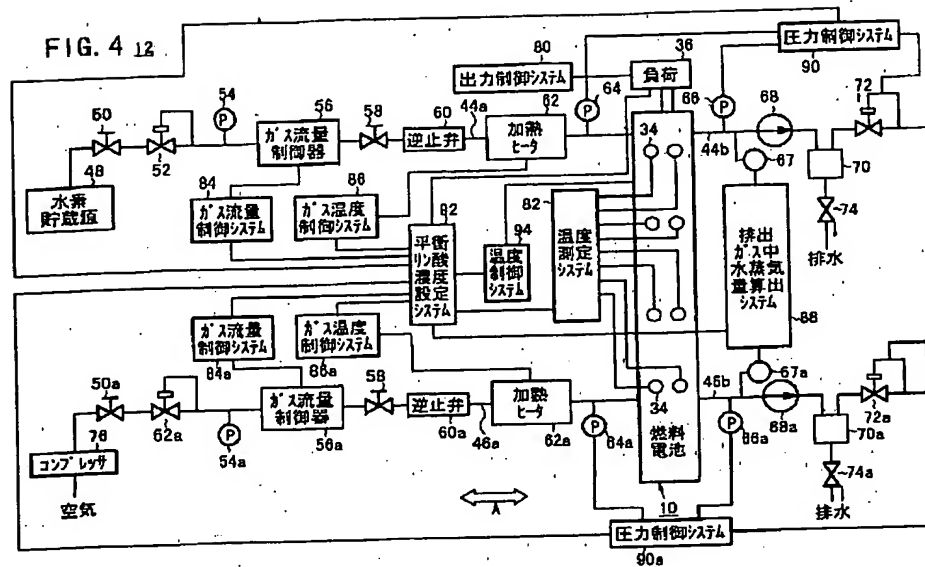
FIG. 3



【図2】



【図 4】



フロントページの続き

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